

Fast approximation for the energy and density of electrons in two dimensions

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(Dated: November 29, 2011)

We derive a self-consistent variant of the energy functional presented in S. Pittalis and E. Räsänen, Phys. Rev. B **80**, 165112 (2009). The resulting scheme is an explicit orbital-free representation of the electron density and energy in terms of the external potential, the number of electrons, and the chemical potential determined upon normalization. The method yields reasonable results for two-dimensional quantum dots, rings, and slabs with a practically negligible computational cost. Both the benefits and drawbacks of the approach are analyzed.

PACS numbers: 71.15.Mb, 31.15.E-, 73.21.La

Orbital-free density-functional theory (OF-DFT) is a computationally appealing method to deal with large systems beyond the reach of conventional DFT. At present, OF-DFT methods allow to deal with systems up to a million atoms.¹ These methods await to be fully explored in the context of low-dimensional systems and nanoelectronic devices. In two-dimensional (2D) physics one of the main challenges of DFT is to deal with regions of the 2D electron gas² comprising hundreds or thousands of interacting electrons, e.g., in the quantum Hall regime. During the recent years successful efforts have been made to describe quantum Hall devices by incorporating the electron-electron (e-e) interactions up to the TF level,³ but improvements regarding, e.g., the exchange-correlation effects neglected in the TF approach are needed.

As the name suggests, OF-DFT is free from the use of the Kohn-Sham orbitals needed in the calculation of the Kohn-Sham kinetic energy, and thus the only explicitly needed variable is the electron density $\rho(\mathbf{r})$. The earliest OF-DFT method can be dated back to the TF theory employing the exact result of the homogeneous electron gas for the kinetic energy, and the Hartree approximation for the e-e interaction. In fact, most orbital-free schemes can be regarded as modifications or improvements to the TF method.⁴ The crux of the problem in OF-DFT is to find an approximation for the (non-interacting) kinetic-energy functional that may be generally applicable. For this ambitious goal, a promising approach is an orbital-free formulation exploiting the potential rather than the density as the basic variable.⁵ As described below, in this work our aim is not to improve over the TF approximation for the kinetic energy.⁶ Instead, our focus is on replacing the Hartree energy with a much simpler expression accounting also for quantum part of the e-e interaction to some extent.

Recently, a 2D orbital-free expression for the energy was shown to lead to a major improvement over the TF results when applied to quantum dots and slabs up to 200 interacting electrons.⁸ However, the energies were not obtained self-consistently, but by using the electron densities from the local-density approximation (LDA).

In this Brief Report, we show that the approach works well also when it is applied fully self-consistently. In doing so, we express the density in terms of the given external potential, number of electrons, and the chemical potential – the latter determined through the particle normalization condition. The functional is tested for a variety of 2D systems including quantum dots, rings, and slabs. A promising overall performance is obtained with an almost negligible computational cost. This gives the method an appealing position in the field of OF-DFT for low-dimensional systems and electronic devices.

In Ref. 8 we showed that using the Gaussian approximation for the *cylindrical* average of the pair density, and a uniform coordinate scaling, the total e-e interaction energy can be expressed as

$$W[\rho(\mathbf{r})] = \frac{\pi}{2} \sqrt{\frac{N-1}{2}} \int d\mathbf{r} \rho^{3/2}(\mathbf{r}), \quad (1)$$

where N is the number of electrons and $\rho(\mathbf{r})$ is the 2D total electron density. In turn, an orbital-free estimation for the total energy can be expressed as

$$E_{\text{tot}}[\rho(\mathbf{r})] = T_{\text{TF}}[\rho(\mathbf{r})] + W[\rho(\mathbf{r})] + \int d\mathbf{r} \rho(\mathbf{r}) v_{\text{ext}}(\mathbf{r}) \quad (2)$$

where

$$T_{\text{TF}}[\rho(\mathbf{r})] = \frac{\pi}{2} \int d\mathbf{r} \rho^2(\mathbf{r}) \quad (3)$$

is the TF kinetic energy in 2D, and the last term is the energy contribution due to the external scalar (confining) potential $v_{\text{ext}}(\mathbf{r})$.

Now, we emphasize a few important points of the energy functional given in Eq. (2). First, for $N = 1$ the functional evidently reduces to the noninteracting TF approximation. Second, for $N \neq 1$ the interaction contribution is a single term that has the simple form of the exchange energy in the LDA but with a different prefactor $\sqrt{N-1}$. In this way, the computational burden of the Hartree term in the TF approximation has been eliminated. On the other hand, $W[\rho]$ in Eq. (1) is evidently not size-consistent as discussed in Ref. 8.

In order to find the ground-state density we have to minimize Eq. (2) for a fixed number of particles. We may first express the total energy in a single integral as

$$E_{\text{tot}}[\rho(\mathbf{r})] = \int d\mathbf{r} F[\rho(\mathbf{r})] = \int d\mathbf{r} \left[\frac{\pi}{2} \rho^2(\mathbf{r}) + \frac{\pi}{2} \sqrt{\frac{N-1}{2}} \rho^{3/2}(\mathbf{r}) + \rho(\mathbf{r}) v_{\text{ext}}(\mathbf{r}) \right]. \quad (4)$$

We have to find a stationary value for the functional $F[\rho(\mathbf{r})]$ with respect to variations in $\rho(\mathbf{r})$. To take the electron number conservation into account, we introduce another functional $G[\rho(\mathbf{r})] = \rho(\mathbf{r})$ so that

$$\int d\mathbf{r} G[\rho(\mathbf{r})] = N. \quad (5)$$

This constraint introduces a Lagrange multiplier μ in the variational equation, which can be written as

$$\frac{dF}{d\rho} - \mu \frac{dG}{d\rho} = 0. \quad (6)$$

Substituting F and G to this equation yields

$$\pi\rho(\mathbf{r}) + \frac{3\pi}{4} \sqrt{\frac{N-1}{2}} \rho^{1/2}(\mathbf{r}) + v_{\text{ext}}(\mathbf{r}) - \mu = 0. \quad (7)$$

As this expression is quadratic in $\rho^{1/2}$, we find an explicit expression for the density,

$$\rho(\mathbf{r}) = \left\{ -\frac{3}{8} \sqrt{\frac{N-1}{2}} + \frac{1}{2} \sqrt{\left[\frac{9}{32}(N-1) - \frac{4}{\pi} [v_{\text{ext}}(\mathbf{r}) - \mu] \right]_+} \right\}^2. \quad (8)$$

This is our key result showing that the density can be solved instantaneously for any external potential v_{ext} and any N . The only variable to be determined numerically is μ that follows from the normalization condition in Eq. (5). The symbol $[\dots]_+$ in Eq. (8) represents an additional constraint that no sign changes under the square in Eq. (8) (leading to unphysical “nodal lines” in the density), nor negative values under the square-root (leading to complex densities), are allowed. Once $\rho(\mathbf{r})$ is determined through Eq. (8), the total energy is obtained from Eq. (2).

Let us emphasize the difference between the present and the TF approximation. In the latter, the variational procedure applied to the total energy leads to an *integral* equation for the density. The TF scheme then transforms into a differential equation (which in 3D leads to the Poisson equation). Instead, our functional is free from this complexity due to the simple expression for the interaction energy [Eq. (1)] in comparison with the Hartree integral utilized by the TF method. Although the Hartree term is simple to calculate in most applications, it may become a bottleneck in very large 2D

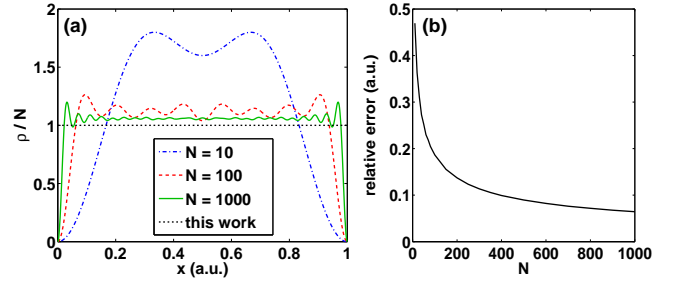


FIG. 1: (color online) (a) Cross section of the electron density in a square quantum well with side length $L = 1$ at $y = 0.5$. The well contains N noninteracting electrons. (b) Relative error in the total energy given by the present functional for the same system as a function of N .

systems. On the other hand, the numerical cost of the present scheme is practically negligible for any N .

Before computational applications we consider some analytic limiting cases. First, as noted above, the interaction energy correctly vanishes at $N = 1$. On the other hand, the density in Eq. (8) becomes simply $\rho_{N=1}(\mathbf{r}) = [\mu - v_{\text{ext}}(\mathbf{r})]_+ / \pi$, i.e., the density profile is the additive inverse of the external potential, and the height of the profile is determined by μ such that the normalization condition is satisfied. As a simple example, for a 2D harmonic oscillator the exact Gaussian-type density is approximated by a parabolic one.

Second, let us consider a 2D square quantum well with side length $L = 1$ and infinite boundaries consisting of N *noninteracting* electrons. Thus, we omit W in Eq. (2) and apply the remaining expression self-consistently. The exact density can be written simply as a sum of a (squared) product of sine (or cosine) functions. In Fig. 1(a) the cross section of the exact density per electron at $y = 1/2$ is compared with the present functional that yields a constant density $\rho = N$, as $v_{\text{ext}}(\mathbf{r}) = 0$ inside the well. Clearly, our approximation for the total density (dashed line with $\rho/N = 1$) improves as a function of N . This favorable property is reflected in the estimation of the total energy: as shown in Fig. 1(b), the energy – here corresponding solely to the TF kinetic energy – becomes more accurate as a function of N in comparison with the exact result, i.e., the sum of the energy eigenvalues.

Next we test the functional for a set of 2D quantum dots, rings, and slabs including the e-e interactions. We use the 2D LDA⁹ in DFT as our reference method; in the range of systems and parameters considered here the LDA has been shown to provide – for the present purpose – sufficiently accurate total energies and densities (see, e.g., Refs. 10 and 11 for quantum dots and rings, respectively). The LDA calculations are performed using the *octopus* code.¹²

Figure 2(a) shows the radial densities obtained from Eq. (8) for 2D harmonic quantum dots defined by $v_{\text{ext}}(r) = \omega^2 r^2 / 2$ with $\omega = 0.5$ (solid lines). The dashed lines show the corresponding LDA results. As expected,

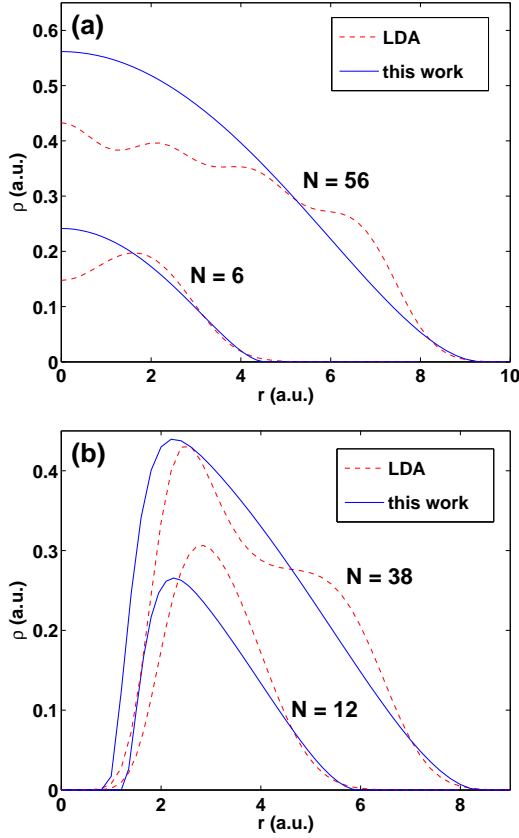


FIG. 2: (color online) (a) Electron densities in harmonic quantum dots containing $N = 6$ and 56 electrons, respectively. The dashed lines denote the conventional density-functional calculations with the local-density approximation. The solid lines correspond to the results of the present orbital-free functional. (b) The same as in (a) but for two quantum rings containing $N = 12$ and 38 electrons, respectively.

the present functional cannot describe the shell structure due to the lack of orbitals. However, the overall shape of the density profile is found in a satisfactory fashion, and, most importantly, the correct radial extent of the density profile is obtained in both cases ($N = 6$ and $N = 56$). Note that the correct density has a Gaussian decay,¹³ whereas in our approximation, by definition, the decay follows the external potential up to the power of one. As an obvious drawback, agreement with the LDA does not seem to improve as a function of N .

In Fig. 2(a) we show the corresponding result for a quantum ring modeled by $v_{\text{ext}}(r) = \omega^2 r^2/2 + V_0 \exp(-r^2/d^2)$ with $\omega = 0.5$, $V_0 = 20$, and $d = 1$. The model potential is the same as the one used in Refs. 11, 14, and 15, the last reference showing direct comparison with an experiment. We find a reasonable qualitative agreement between the present functional and the LDA. The qualitative agreement is similar for both $N = 12$ and $N = 38$.

Apart from densities, it is important to assess the performance of the present functional for total energies. In

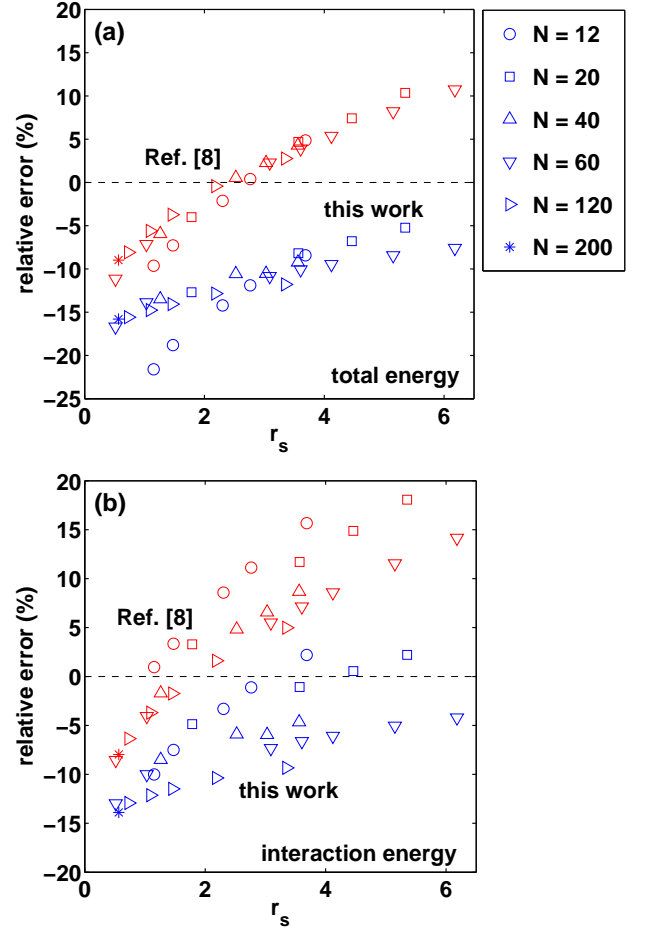


FIG. 3: (color online) (a) Relative error in the total energy (with respect to the local-density approximation) for rectangular quantum slabs with $N = 12 \dots 200$ as a function of the density parameter r_s . The results of the present functional and that of Ref. 8 – employing a non-self-consistent version of the present functional – have been indicated. (b) The same as (a) but for the electron-electron interaction energy.

Ref. 8 this was done non-self-consistently by applying the LDA densities as an input for the total-energy functional in Eq. (2). Here we use the present self-consistent approach to the same extensive set⁸ of rectangular quantum slabs.^{16,17} The external potential is defined by steep boundaries, so that the side-length ratio is fixed to two, and the shorter side is varied between 5 and 60 a.u. The number of electrons is varied in the range $N = 12 \dots 200$.

In Fig. 3(a) we show the relative total-energy differences from the reference LDA results for a set of rectangular quantum slabs as a function of the average electron density in terms of the Wigner-Seitz radius,² $r_s = \sqrt{A/(\pi N)}$, where A is the area of the slab. Results are shown for the present functional and for the non-self-consistent calculation of Ref. 8. The latter scheme seems to outperform the former at $r_s \gtrsim 4$ when the average density becomes low. This improvement is accidental in the sense that the self-consistent density of our functional

[Eq. (8)] remains *constant* in the slab regardless of N and r_s , as v_{ext} is zero inside the boundaries. As a function of N , however, it can be expected that the present functional becomes more accurate, since the actual density becomes flatter in the large- N limit – similarly to the noninteracting case analyzed within Fig. 1 above.

Figure 3(b) shows the relative errors in the interaction energy W in Eq. (1) that can be compared to the LDA result, $W_{\text{LDA}} = E_H + E_{xc}$, where xc refers to the exchange and correlation. The results are similar to those for the total energy, though now the present self-consistent functional seems to perform slightly better than the non-self-consistent one.

Quantum slabs as defined above are also useful model systems in the analytic and self-consistent application of our approximation in limiting situations. In contrast with the analysis above (Fig. 1) we now include the e-e interactions. First, when the area A of the slab is fixed and N goes to infinity we get $\mu \approx [\pi/A + 3/(4\sqrt{2})] N$ and $E_{\text{tot}} \approx (\mu/2 + N/8)N$. Then, when $N \gg 1$ but finite and A goes to infinity, we get $\mu \approx 3\pi N/(4\sqrt{2}A)$ and $E_{\text{tot}} \approx 2\mu N/3$. Finally, when both N and A approach infinity while keeping the (self-consistent) density constant, we get, again, $\mu \approx 3\pi N/(4\sqrt{2}A)$ and $E_{\text{tot}} \approx 2\mu N/3$. In all these three limiting cases, our approximation apparently *fails* to satisfy the fundamental relation $\mu = dE_{\text{tot}}/dN$. We attribute this fact to the fact

that W is not size-consistent.⁸ In our preliminary tests we have not found severe consequences of this deficiency: in quantum dots, for example, the relative errors in energy *differences* $E(N) - E(N-1)$, when compared to the LDA calculations, are of the same order as the relative errors in the total energies themselves.

Despite the obvious formal deficiencies, the present approach has a few important advantages. First, the approximation is extremely fast in the sense that no iterations apart from the numerically trivial determination of μ are required. Hence, the functional could be used as the first approximation for the electron density in the conventional Kohn-Sham scheme. Secondly, despite the remarkable simplicity the functional yields reasonable magnitudes for the total and interaction energies. Hence, the present analysis supports further improvements and extensions in developing orbital-free methods for large-scale two-dimensional electronic devices.

Acknowledgments

This work was supported by the Academy of Finland (E.R. and G.B.), Wihuri Foundation (E.R.), ERASMUS Internship Programme (G.B.), and the DOE grant DE-FG02-05ER46203 (S.P.). CSC Scientific Computing Ltd. is acknowledged for computational resources.

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⁶ It has been observed that the TF approximation for the kinetic energy works remarkably well for systems considered here⁷. See also Fig. 1.

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